

Interactive comment on “4-D comparison method to study the NO_y partitioning in summer polar stratosphere – Influence of aerosol burden” by G. Dufour et al.

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General comments

Understanding the budget and partitioning of the No_y family is a key to the quantitative understanding of stratospheric ozone loss. Despite No_y chemistry has been studied for more than two decades, some area of uncertainties remain especially regarding the ability of models to reproduce observed distributions in the lower stratosphere. This paper presents an interesting case study where profile measurements of several components of the No_y family, simultaneously recorded in the summer polar strato-

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sphere from balloon borne FTIR and DOAS experiments, are used to test state-of-the-art model calculations of the No_x/No_y partitioning. Results stress (1) the importance of accounting for the spatial variability of the measured trace gases when performing the inter-comparisons, and (2) the role of aerosols in controlling the No_x/No_y ratio below 20 km of altitude. It is found that the observed No_y partitioning can be well reproduced using the most recent reaction rate coefficients as recommended in JPL-2003.

In this exercise, the investigators have adopted a rather sophisticated inter-comparison methodology, based on a combination of 3D CTM and 1D photochemical calculations, the relevance of which is convincingly introduced and supported by the results. However I found the detailed description of the procedure unclear with respect to the following two questions:

(1) One key point of the paper is the need to account for spatial variability effects, as illustrated in Figure 2. The spatial variability is likely to be driven by dynamics and/or photochemical variations. Reading through the text, it is very difficult to judge whether the adopted methodology accounts for both transport and photochemical effects, or only for the latter. E.g. it is mentioned in the first paragraph of section 3 that the 3D simulations were initialized on 15 Oct. 2000 and driven by ECMWF analysis until June 2001, while the comparison is made on 21–22 August. Is the dynamics of June also adequate for August? Also the correction formula presented in Equation 2 seems to only account for photochemical changes between the actual measurement time and the time of the 3D CTM output. If I am correct, any change due to transport effect during that period will not be corrected for, which should be acknowledged by the authors.

(2) The discussion of the aerosol effect is shortly evacuated in a single paragraph at the end of the paper, although it is presented as a key result of the paper both in the title and in the abstract. Basically the authors argue that the aerosol distribution used by the model is not “realistic” enough, and that much better results can be obtained using aerosol surface areas measured in 2002. However they also stress the large variability and uncertainty of these measurements, so that the reader may wonder whether these

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data are really appropriate in the present context. It would help a lot to show how different are measured and model aerosol profiles, possibly using a Figure. Another question not answered is: how large is the variability of the measured aerosol content in comparison to the original “unrealistic” profiles? If paper length is an issue, I might recommend to remove Table 1 and 2, which are not absolutely needed in the present paper.

Besides the above considerations, I found the paper generally well written and properly structured, with proper credit to existing literature. Figures are clearly drawn and captions informative enough. Altogether, I think this paper provides interesting and useful results, which certainly deserve to be published in ACP. However some adjustments to the text (and possibly one additional Figure) are needed, to answer to two main comments above. There are also a few additional comments (mostly of editorial nature), which are given below.

Specific comments

- Page 8173, line 25: replace “within” by “in”
- Page 8174, line 2: correct “Using this updated rate...” by “Using these updated rate...”
- Page 8174, line 8: the reference to Sander et al., 2003 is not given at the end of the paper (only one to Sander et al., 2002).
- Page 8174, same line: as written, the sentence beginning with “A robust” is not correct. I'd suggest to remove the first “inter-comparison” to “A robust initialization scheme is used to inter-compare observed and simulated profiles...”
- Page 8174, line 10: remove “Here” in the sentence beginning after “sunset”.
- Page 8174, line 23: remove “The ozone profile was also used in this study”.
- Page 8174, line 26: change “the consistency of O₃ and NO₂ retrieved vertical profiles” by “the consistency of the retrieved O₃ and NO₂ vertical profiles”

- Page 8176, line 15: change "...between 89° and 91° solar zenith angle" by "...between 89° and 91° of solar zenith angle"

- Page 8177, line 17: replace "22 August 2002" by "22 August 2001"

- Page 8177, line 21: remove the word "retrieved" after the list of molecules

- Page 8177, line 23: I think that the sentence beginning with "The disagreement for HNO_3 is explained by..." should be moved downward so that it comes as a conclusion of the discussion of the results presented in Figure 4.

- More fundamentally, it is clear from the paper that No_y is underestimated by Reprobus under the flight conditions (polar summer). There is no comment on the possible reasons for this misbehavior of the model. Is this a general problem of all 3D CTM or is this specific to Reprobus? If possible, the paper would gain in interest by further developing this aspect in the discussion.

- Page 8180, line 1: replace "...this discrepancy can be explained best by..." by "...this discrepancy can be explained to a large extent by..."

- Page 8180, line 8: correct "assigns" by "assign"

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